Supporting Information

Light-Assisted Anti-Wrinkling on Azobenzene-Containing Polyblend Films

Juanjuan Wang,[1,#] Haipeng Ji,[2,#] Yanqian Guo[1], Bin Wang,[3] Xue Han,[1] Lele Li,[4] Fuqi Wu,[3] Jingqing Li,[4,]* and Conghua Lu [1,]*

[1] School of Materials Science and Engineering, Tianjin Key Laboratory of Building Green Functional Materials, Tianjin Chengjian University, Tianjin 300384, China.
*E-mail: chlu@tju.edu.cn

[2] No 46 Institute, China Aerospace Science and Industry Corporation Sixth Academy, Huhhot 010010, China.

[3] School of Science, Tianjin Chengjian University, Tianjin 300384, China.

[4] School of Materials Science and Engineering, Tianjin University, Tianjin 300072, China.
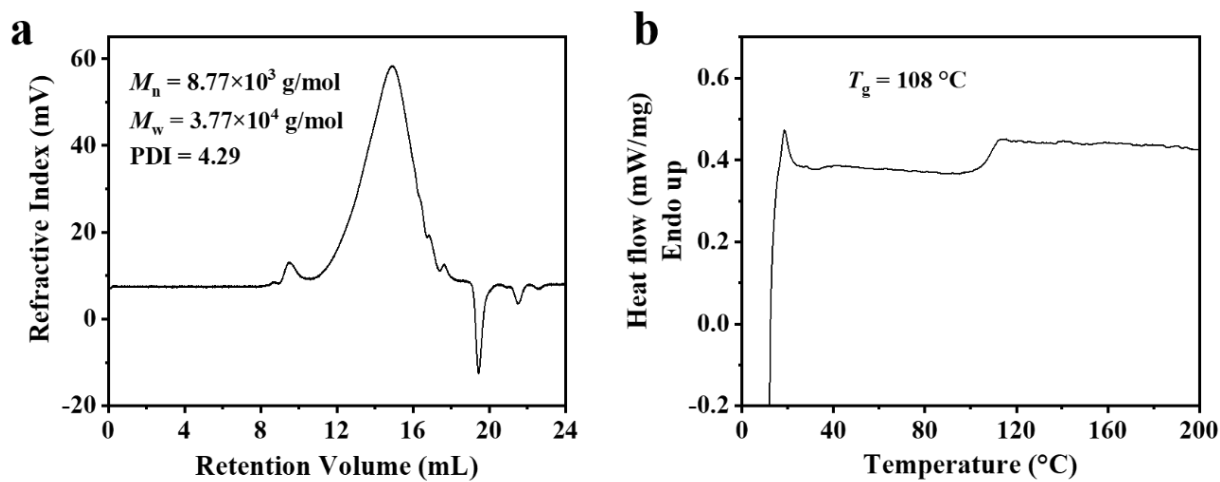*E-mail: jqli11103@tju.edu.cn

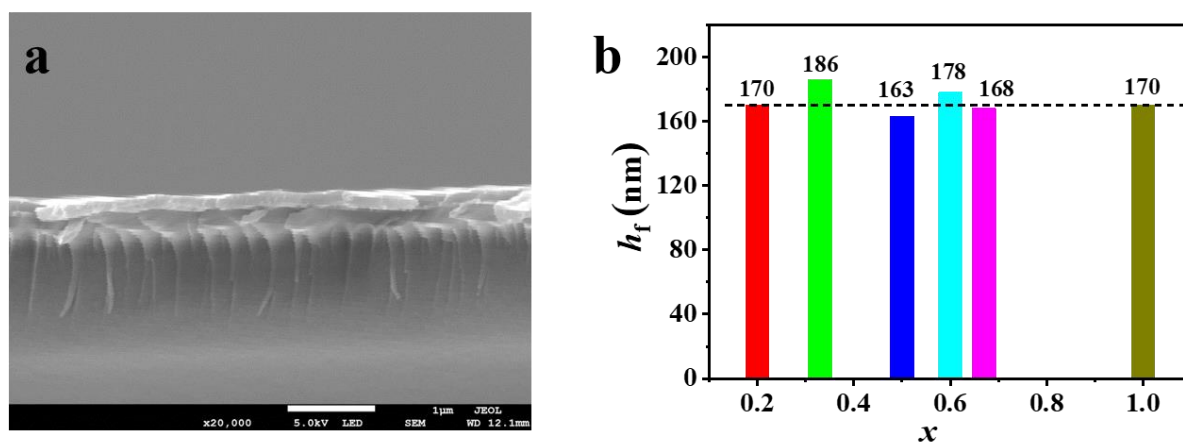**Figure S1.** The GPC (a) and DSC (b) curves of the obtained PAAB.

Figure S2. (a) Typical SEM image of the cross-section of the $PAAB_x$-$PMMA_{1-x}$ film when $x = 0.6$. (b) The relation of the thickness (h_f) of $PAAB_x$-$PMMA_{1-x}$ films with x.
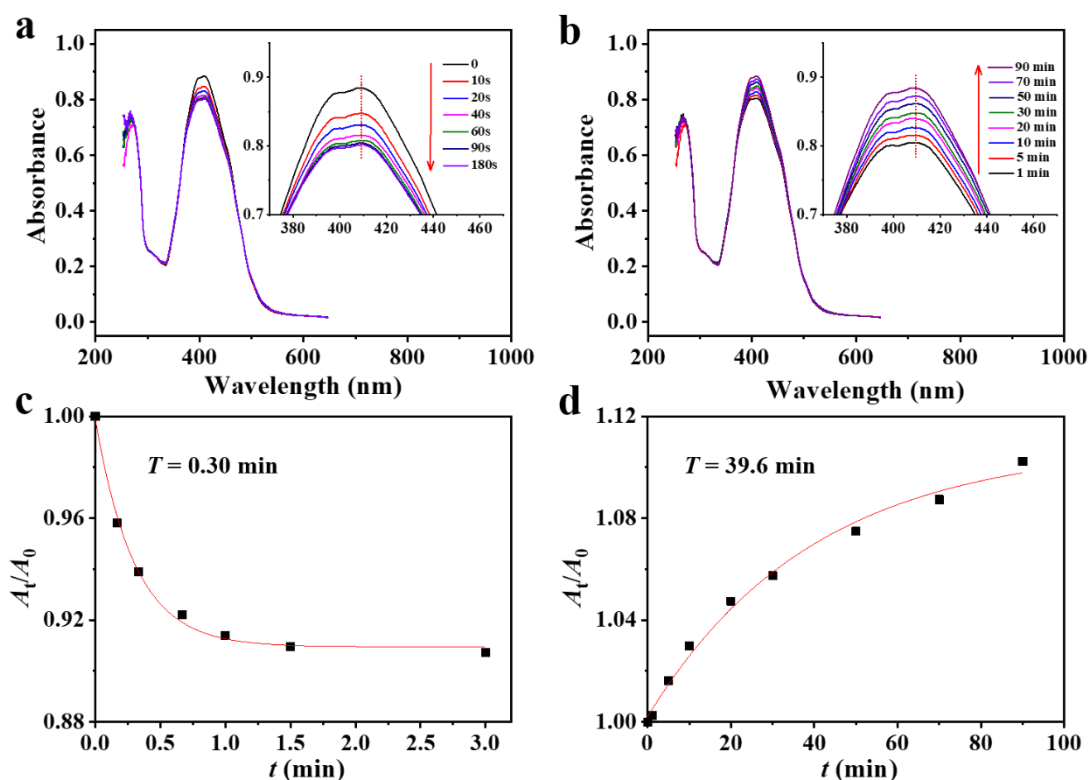
Figure S3. UV-Vis absorption spectrum evolution (a, b) and the relationship between the relative absorbance A_t/A_0 and time t (c, d) of the PAAB film with visible light irradiation at $I=5$ mW/cm^2 (a, c) and its recovery in darkness (b, d). Inset in (a,b) is the corresponding spectrum magnification around the λ_{max}. The solid line in (c,d) is the corresponding fitting curve.

The photoisomerization of azo-moieties is sensitive to the local environment surrounding the moieties, which can be evaluated by measuring the isomerization rate.[1] From Figure S3a,b, the absorbance at λ_{max} before the light irradiation (A_0) and after the irradiation for different time (A_t) can be obtained. The relative absorbance (A_t/A_0) of the samples represents the relative amount of the *trans* isomers remaining at the time t. Furthermore, the variations of A_t/A_0 with t indicate the kinetics of the photoisomerization. For Figure S3c,d, the variations can be best fitted by the first-order exponential decay function

$$A_t/A_0 = A' + A'' \exp(-t/T) \tag{S1}$$

where T is the characteristic time of the decay process. The R-Square (COD) values in the above fittings are >0.99. T obtained from the curve fitting is 0.30 min and 39.6 min for Figure S3c,d, respectively.
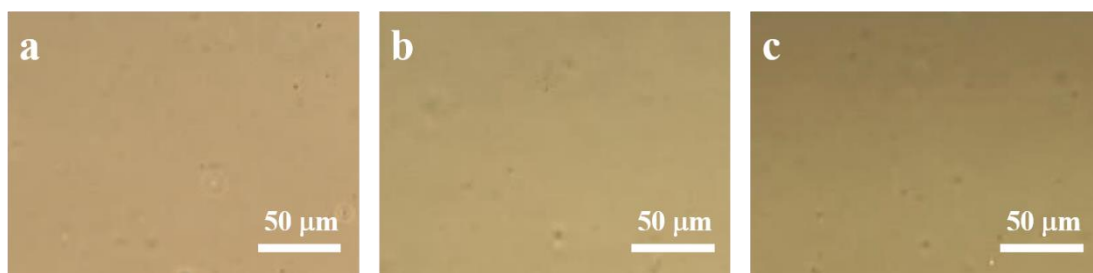
Figure S4. Optical images of as-prepared $PAAB_x$-$PMMA_{1-x}$ films on the PDMS substrate with $x = 0.20$ (a), 0.50 (b), 0.67 (c), respectively.
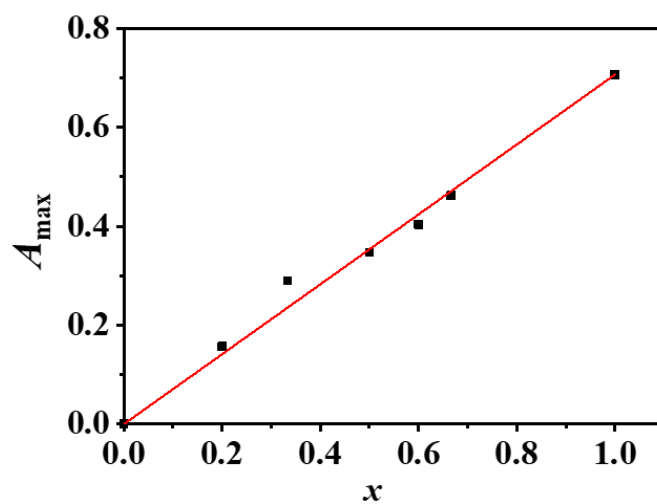

Figure S5. The dependence of A_{max} on x. The solid line is from the linear regression of data pairs.
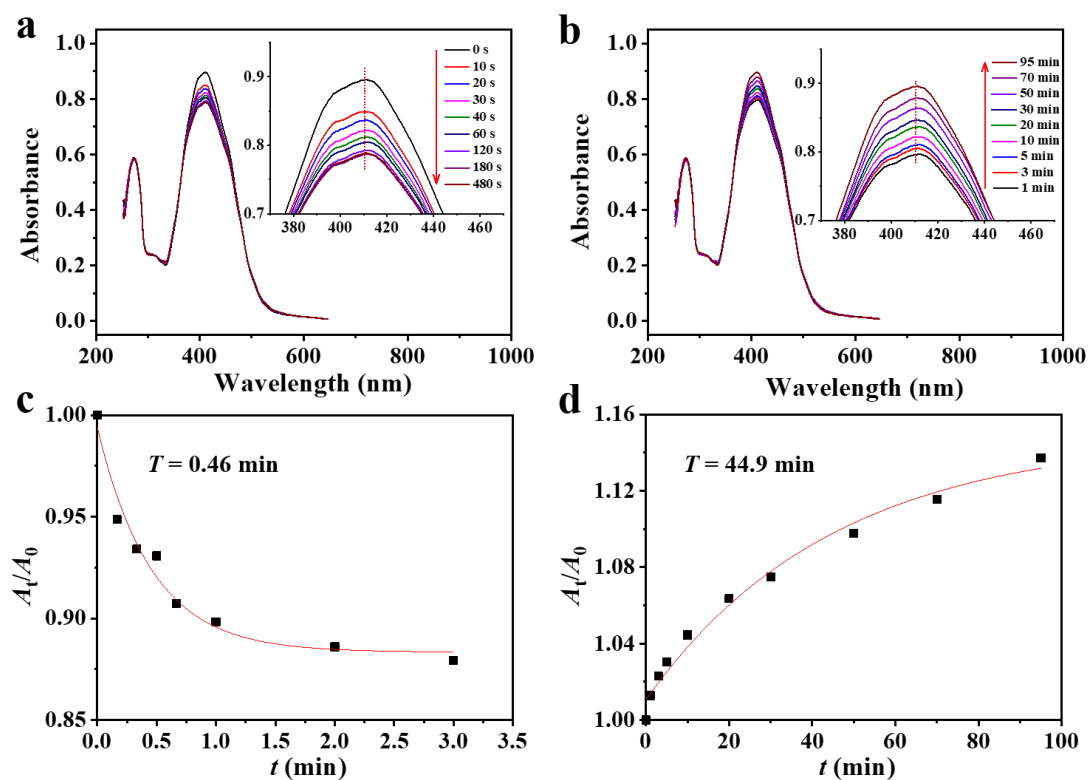
Figure S6. UV-Vis absorption spectrum evolution (a, b) and the relationship between the relative absorbance A_t/A_0 and time t (c, d) of the PAAB$_{0.5}$-PMMA$_{0.5}$ film with visible light irradiation at $I=5$ mW/cm^2 (a, c) and its recovery in darkness (b, d). The inset in (a,b) is the corresponding spectrum magnification around the λ_{max}. The solid line in (c,d) is the corresponding fitting curve.

For Figure S6c,d, the variations of A_t/A_0 with t can also be best fitted by the first-order exponential decay function (Equation (S1)), where both COD values are >0.97. T obtained from the curve fitting is 0.46 min and 44.9 min for Figure S6c,d, respectively. Comparing with the PAAB film, the photo-induced *trans→cis* isomerization and thermal-induced *cis→trans* isomerization rate decline for the PAAB$_{0.5}$-PMMA$_{0.5}$ film.
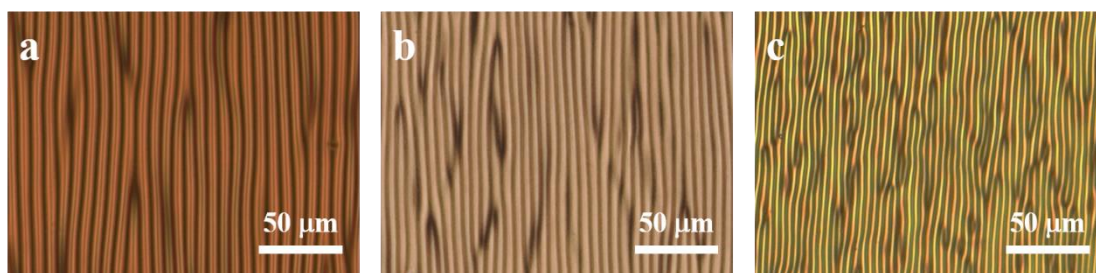
Figure S7. Optical images of the (PAAB$_{0.5}$-PMMA$_{0.5}$)/PDMS bilayer after releasing prestrain with $\varepsilon =$ 10% during visible light irradiation at $i = 0$ (a), 3 (b), 12 (c) MPa, respectively.
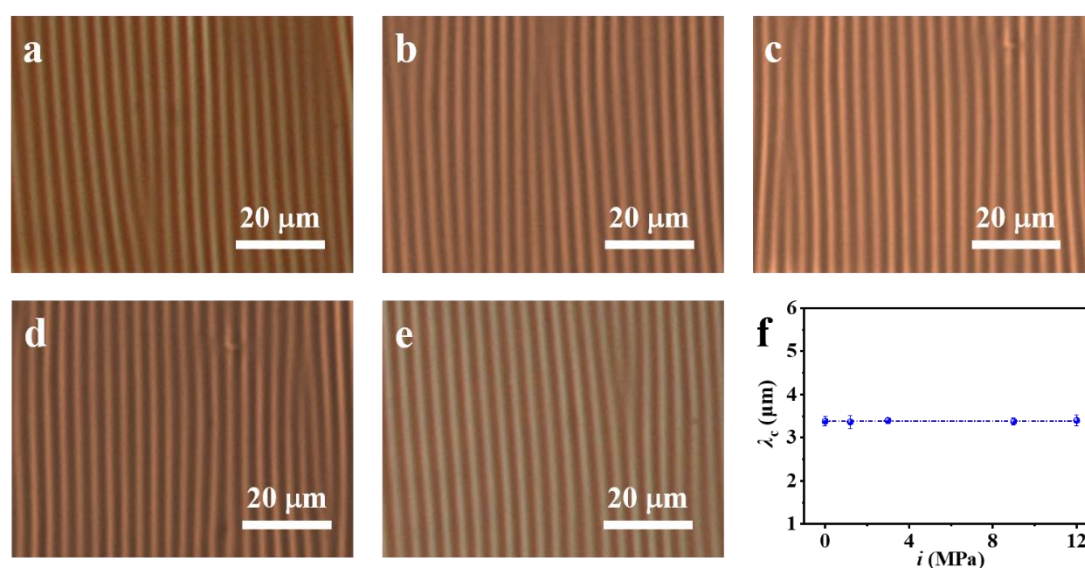

Figure S8. (a-e) Optical images of the PMMA/PDMS bilayer after releasing prestrain with $\varepsilon = 2\%$ during the visible light irradiation at $i = 0$ (a), 1.2 (b), 3 (c), 9 (d) and 12 (e) MPa, respectively. (f) Plots of λ_c as a function of i.
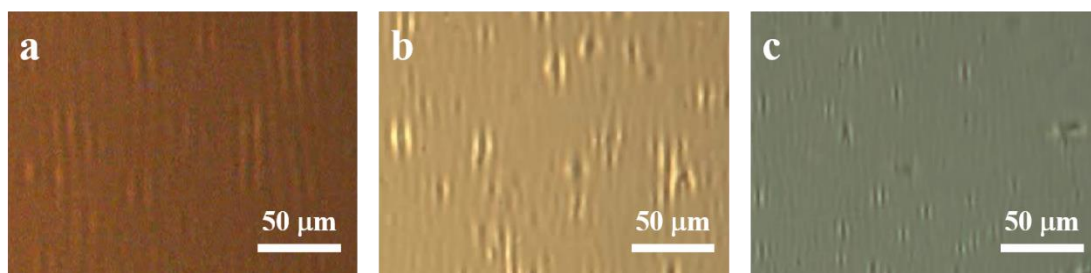
Figure S9. Optical images of the critical wrinkling $PAAB_{0.5}$-$PMMA_{0.5}$ film on the PDMS substrate with i: 0 (a), 3 (b), 12 (c) MPa, respectively.
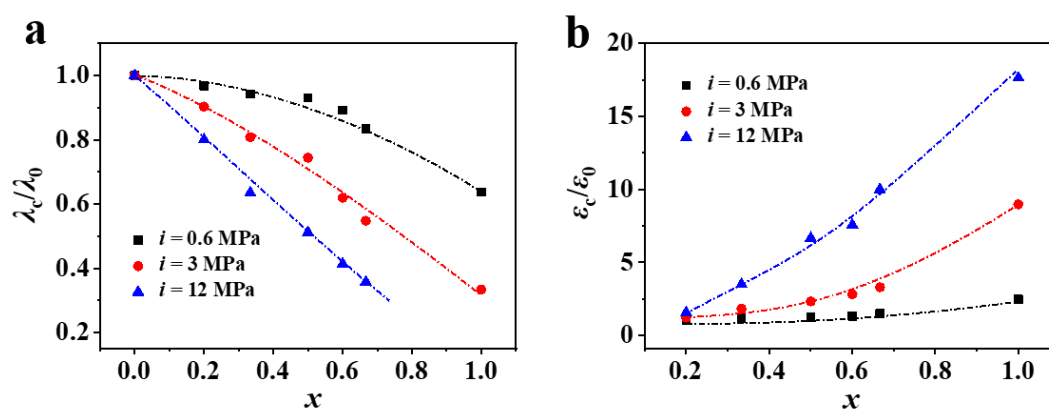

Figure S10. The dependence of λ_c/λ_0 (a) and $\varepsilon_c/\varepsilon_0$ (b) on x in the $PAAB_x$-$PMMA_{1-x}$ films under different experiment condition i employed. The dashed lines were drawn to visualize the general trend.

REFERENCES

1. Y. Deng, Y. Li, X. Wang, Colloidal sphere formation, H-aggregation, and photoresponsive properties of an amphiphilic random copolymer bearing branched azo side chains. *Macromolecules* **2006,** *39,* 6590-6598.